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14. ABSTRACT
Attosecond hole dynamics: While studying the spectrum of the high harmonics produced from CO2, we discovered that multiple orbitals can ionize at the same time. This was an unexpected result suggesting that tunneling often launches attosecond dynamics. This research, including an image of the wave packet motion, was published in Nature [2].

Imaging orbitals via tunneling: To gain additional insight into the ionization of lower orbitals, we measured the ionization probability as a function of direction. Using H_2 (because of its simplicity) we showed that the tunnel ionization rate essentially follows the orbital structure (Physical Review Letters [3]). Then, we turned to HCl and measured the relative probability of tunnel ionizing the HOMO and the HOMO-1 orbital (Science [1]). Finally we developed an electron tomographic imaging technique [6].

15. SUBJECT TERMS
ATTOSECOND,Molecular Orbitals, Imaging, High harmonics, tunneling, ionization

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Annual Accomplishments:

Our objective for the whole AFOSR proposal was to image molecular orbitals with pi symmetry. Below we outline our accomplishments during 2009:

Attosecond hole dynamics: While studying the spectrum of the high harmonics produced from CO_2 we uncovered that multiple orbitals were ionizing at the same time when the molecule was viewed from some directions. This was an unexpected and exciting result because it implied that a hole wave packet was launched in the molecule. We measured this wave packet which, in CO_2, moved with a natural time scale of ~ 1 fs. This research, suggesting that tunneling often launches previously unsuspected attosecond dynamics in molecules, was published in *Nature* as a full article [2].

Imaging orbitals via tunneling: To gain additional insight into the ionization of lower orbitals we measured the ionization probability as a function of direction in two molecules. We named this new orbital imaging method a "molecular STM" because of its parallel with a conventional "scanning tunneling microscope". Using H_2 (because of its simplicity) in 2008 we showed that the tunnel ionization rate in 800 nm light varies as a function of angle essentially following the orbital structure. This work was published during 2009 in *Physical Review Letters* [3]. Exploiting this fact, we turned to HCl, and Confirmed that not only the HOMO, but also the HOMO-1 ionized. This work was published in *Science* [1]. Finally we improved experimental methods for measuring tunneled electrons by developing an electron tomographic imaging technique [6].

Infrared experiments: Tunneling is a better approximation for ionization if done with infrared radiation. Orbital tomography is also preferentially performed with infrared radiation because the recollision electron wave length is shorter. In a paper published in *Physical Review Letters* we studied the wavelength scaling of high harmonic generation using infrared radiation [4]. As the year drew to an end we extended our infrared experiments to more complex molecules [5].

Publications during 2009:

1. H. Akagi, T. Otobe, A. Staudte, A. Shiner, F. Turner, R. Dorner, D.M. Villeneuve and P.B. Corkum, "Direct Observation of Tunneling from HOMO-1 in HCl", *Science*, **325**, 1364 (2009).

2. O. Smirnova, Y. Mairesse, S. Patchkovskii, N. Dudovich, D. M. Villeneuve, P. B. Corkum, and M. Y. Ivanov, "Probing Inner Orbitals and Electron Tunneling in Molecules with High Harmonic Interferometer", *Nature*, **460**, 972 (2009).

3. A. Staudte, S. Patchkovskii, D. Pavicic, H. Akagi, O. Smirnova, D. Zeidler, M. Meckel, D.M. Villeneuve, R. Dörner, M. Ivanov and P.B. Corkum, "Angular Tunneling Ionization Probability of H_2" *Phys. Rev. Lett.* **102**, 033004 (2009).

4. A. D. Shiner, C. Trallero-Herrero, N. Kajumba, H.-C. Bandulet, D. Comtois, F. Legare, M. Gigeure, J-C. Kieffer, P. B. Corkum, and D. M. Villeneuve, "Wavelength Scaling of High Harmonic Generation Efficiency", *Phys Rev Lett.* **103**, 073902 (2009).

5. C. Trallero-Herrero, B. E. Schmidt, A. D. Shiner, Philippe Lassonde, ´Eric Bisson, J-C. Kieffer, P. B. Corkum, D. M. Villeneuve, F. Legare, "High harmonic generation in Ethylene with infrared pulses", Chemical Physics, **366,** 33 (2009).

6. C. Smeenk, L. Arissian, A Staudte, D. M. Villeneuve, and P. B. Corkum, "Tomographic imaging of charged particles", J. Phys. B. **42**, 185402 (2009).

Changes to objectives: None

New program manager:

Extensions: None:

Final report: